

Nonequilibrium conductance of asymmetric nanodevices in the Kondo regime

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The scaling properties of the conductance of a Kondo impurity connected to two leads that are in or out of equilibrium has been extensively studied, both experimentally and theoretically. From these studies, a consensus has emerged regarding the analytic expression of the scaling function. The question addressed in this brief report concerns the properties of the experimentally measurable coefficient α present in the term describing the leading dependence of the conductance on eV/T_K , where V is the source-drain voltage and T_K the Kondo temperature. We study the dependence of α on the ratio of the lead-dot couplings for the particle-hole symmetric Anderson model and find that this dependence disappears in the strong coupling Kondo regime in which the charge fluctuations of the impurity vanish.

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Universality of scaling plays a central role in the Kondo effect¹ which describes the interaction of a magnetic impurity with conduction electrons. As the temperature T is lowered below a crossover scale, denoted by the Kondo temperature T_K , the impurity spin becomes screened by conduction electrons. Recent advances in nanofabrication techniques now allow for the experimental exploration of Kondo physics by attaching two conducting leads, which we denote left L and right R, to a smaller region, such as a semiconductor quantum dot (QD) or a single molecule transistor (SMT), each of which may act as an effective impurity spin. One typically applies a voltage difference V between the leads and measures the differential conductance $G(T, V) = dI/dV$ which grows at low T and V due to Kondo correlations. This behavior is expected to be described by a scaling form $G(T, V) = G_0 F(T/T_K, eV/T_K)$ where $G_0 = G(0, 0)$ and F is a universal scaling function. The scale T_K can vary between 100mK in QD devices^{2,3} and 150K in SMTs.^{4,5}

Such devices are more accurately described by the Anderson model (AM) which takes into account charge fluctuations of the impurity. The Kondo model (KM) is recovered from the particle-hole symmetric AM in the limit $U \rightarrow \infty$ where U is the charging energy in the AM. For both models, for low energies $T, eV \ll T_K$ the leading corrections to the conductance are given by (up to a redefinition of T_K)

$$F(T/T_K, eV/T_K) = 1 - c_T \left(\frac{T}{T_K}\right)^2 - \alpha c_T \left(\frac{eV}{T_K}\right)^2 + \dots, \quad (1)$$

with various values predicted for the coefficient α from the AM⁶⁻⁹ and Kondo model KM.¹⁰⁻¹³ Recently, α was measured by two experiments, one done by Grobis *et al.* on a quantum dot device¹⁴ and the other done by Scott *et al.* on an ensemble of single molecule transistors,¹⁵ where the QD and SMTs were tuned to the Kondo regime $T, eV \ll T_K$. In the QD experiment, T_K varied from 150 to 300mK by varying the gate voltage in a single device, and a value $\alpha_{\text{QD}} = 0.1 \pm 0.015$ was measured. In the SMTs experiment, T_K ranged from 34 to 155K in 29 different devices, and α showed a systematic deviation from the QD value, $\alpha_{\text{SMT}} = 0.051 \pm 0.01$ (see Refs. 14,15

for the precise fitting range). Various possible explanations were pointed out for the systematic difference of α in SMTs. Among those, the relative asymmetry of the L and R coupling [denoted by A in Eq. (4)] was considered as a relevant issue.¹⁵

With this experimental motivation, we calculate α for arbitrary device asymmetry. We consider the particle-hole symmetric Anderson model (SAM) which generically includes charge fluctuations in the impurity. We find that α is independent of the degree of L-R asymmetry only in the Kondo limit. Once charge fluctuations are included, there is a dependence of α on the L-R asymmetry. We compare our result to previous theoretical results and also comment on the relevancy to the experiments. We find that the low value of α measured in SMTs can not be accounted for within the symmetric Anderson model.

Our phenomenological approach consists of a modification of Nozières Fermi liquid (FL) theory¹⁶ to account for charge fluctuations in the SAM. We pay special attention to the effect of shifting the Kondo resonance at finite voltage [see Eq. (15)]. Our result is a generalization of Oguri's calculation of the conductance which used a non-perturbative result for the Green's function of the SAM,⁸ and is found to reduce to Oguri's result for the special case he considered.

The model of a single Anderson impurity connected to L and R leads is

$$H = H_0 + H_d + H_t, \quad (2)$$

$$H_0 = \sum_{k\sigma} \sum_{i=L,R} \epsilon_k c_{k\sigma i}^\dagger c_{k\sigma i},$$

$$H_d = \epsilon_d \sum_{\sigma} d_{\sigma}^\dagger d_{\sigma} + U d_{\uparrow}^\dagger d_{\uparrow} d_{\downarrow}^\dagger d_{\downarrow},$$

$$H_t = \sum_{k\sigma} \sum_{i=L,R} v_i (d_{\sigma}^\dagger c_{k\sigma i} + \text{H.c.}), \quad (3)$$

where d_{σ} annihilates an electron with spin σ in the quantum dot d -level, $c_{k\sigma i}$ annihilates a conduction electron with momentum k and spin σ in the $i = L, R$ lead,

$\epsilon_k = \hbar v_F k$, and v_F is the Fermi velocity. In the SAM that we will consider here, $\epsilon_d = -U/2$. It is convenient to define the L-R asymmetry parameter

$$\text{L-R asymmetry : } A = \frac{4\Gamma_L\Gamma_R}{(\Gamma_L + \Gamma_R)^2}. \quad (4)$$

The chemical potentials μ_i ($i = L, R$), satisfying $\mu_L - \mu_R = eV$, are measured relative to the Fermi level defined at zero voltage. Then the ratio

$$B = -\mu_L/\mu_R, \quad (5)$$

describes the relative voltage drop across the L and R tunnel junctions which could depend on the capacitive couplings of the leads and QD or SMT and which we treat as a second L-R asymmetry parameter.

One can define the retarded d Green's function as $G^R(\epsilon) = -i \int_0^\infty dt e^{i\epsilon t} \langle d(t) d^\dagger + d^\dagger d(t) \rangle$. With these definitions, the current is given by the Meir-Wingreen formula¹⁷

$$I = \frac{2e}{h} A \int_{-\infty}^{\infty} d\epsilon [f_L - f_R] \Delta [-\text{Im} G^R(\epsilon)], \quad (6)$$

with $f_i = f(\epsilon - \mu_i)$ ($i = L, R$), $f(\epsilon) = [1 + \exp(\epsilon/T)]^{-1}$, $\Delta = \Gamma_L + \Gamma_R$, $\Gamma_i = \pi \nu v_i^2$ ($i = L, R$), and ν is the density of states in the leads.

Below, we formulate an effective theory for the SAM that allows us to obtain the Green's function $G^R(\epsilon)$ for $\epsilon, T, eV \ll \tilde{\Delta}$ [$\tilde{\Delta}$ is the characteristic energy scale defined more explicitly below Eq. (8)]. In the Kondo limit $\tilde{\Delta} \rightarrow T_K$. We define linear combinations of the annihilation operators for the L and R leads

$$a_{k\sigma}^{(s)} = \frac{v_L c_{k\sigma L} + v_R c_{k\sigma R}}{v}, \quad a_{k\sigma}^{(p)} = \frac{-v_R c_{k\sigma L} + v_L c_{k\sigma R}}{v}, \quad (7)$$

where $v = \sqrt{|v_L|^2 + |v_R|^2}$. Only the s -wave particles are coupled to the d -level, since $H_t = \sum_{k\sigma} v d_{\sigma}^\dagger a_{k\sigma}^{(s)} + \text{H.c.}$

The notion of a local Fermi liquid (FL), due to Nozières,¹⁶ was originally applied to the Kondo model but is actually more general and can be applied to the AM and, in particular, to the less complicated SAM. The quasiparticles of this FL theory are simply scattering states whose incoming part coincides with that of the s -wave particles $a_{k\sigma}^{(s)}$ [the precise definition is given after Eq. (10)]. The theory itself consists of a low energy expansion of their scattering phase shift as a function of energy ϵ (measured from the Fermi level) and of quasiparticle density n_σ ,

$$\delta_\sigma = \delta_0 + \frac{\epsilon}{\tilde{\Delta}} - \frac{\beta n_\sigma}{\nu \tilde{\Delta}} + \dots, \quad (8)$$

where $\tilde{\uparrow} = \downarrow$, $\tilde{\downarrow} = \uparrow$, $\tilde{\Delta}$ is the energy scale over which the phase shift varies in the SAM, and β is a coefficient to be determined. For the general AM, the scattering phase shift at the Fermi energy δ_0 can be extracted using the

Friedel sum rule¹⁸ $\delta_0 = \text{Im}[\ln G^R(0)|_{T=eV=0}] - \pi$ combined with exact results for G^R . In the SAM, particle-hole symmetry and the adiabatic connection to the $U = 0$ case implies $\delta_0 = \pi/2$.

The Wilson ratio $R = (\delta\chi/\chi)/(\delta C_v/C_v)$ is the ratio between the relative impurity contribution to the susceptibility and to the specific heat. It can be calculated from the phase shift expansion, Eq. (8), to be¹⁶

$$R = 1 + \beta. \quad (9)$$

We will use this equation to determine β in terms of R which is a parameter describing the amount of charge fluctuations in the SAM.

As an equivalent way to determine β , consider an enhancement of the Fermi energy by an amount ϵ by adding to the Fermi sea a density of quasiparticles $n_\sigma = (\nu\epsilon)$. For the KM, Nozières argued that at energy ϵ , corresponding to the new Fermi energy, one has $\delta_\sigma = \delta_0$ since the Kondo resonance is tied to the Fermi level. Using Eq. (8) and $n_{\tilde{\sigma}} = \nu\epsilon$, this implies $\beta = 1$. This argument should be modified for the AM: a shift of the Fermi level by this transformation also implies that $\epsilon_d \rightarrow \epsilon_d - \epsilon$, as measured relative to the new Fermi level at $+\epsilon$. Therefore a finite amount of charge $e\delta n_d$ enters into the d -level which, for small ϵ , is given in terms of the charge susceptibility $\delta n_d = -\frac{dn_d}{d\epsilon_d}\epsilon$. Here $n_d = \sum_\sigma \langle d_\sigma^\dagger d_\sigma \rangle$. Due to the Friedel sum rule,¹⁸ the phase shift at the new Fermi energy after this transformation is different than δ_0 and given by $\delta_\sigma = \pi n_d/2 = \delta_0 - (\pi/2)\frac{dn_d}{d\epsilon_d}\epsilon$. Using Eq. (8) and $n_{\tilde{\sigma}} = \nu\epsilon$ implies $\beta = 1 + \frac{\pi\tilde{\Delta}}{2}\frac{dn_d}{d\epsilon_d}$.

This phase shift expansion can be equivalently described by a Hamiltonian

$$\begin{aligned} H &= H_0(a^{(p)}) + H_0(b) + \delta H, \\ \delta H &= -\frac{1}{2\pi\nu\tilde{\Delta}} \sum_{kk'\sigma} (\epsilon_k + \epsilon_{k'}) b_{k\sigma}^\dagger b_{k'\sigma} \\ &\quad + \frac{R-1}{\pi\nu^2\tilde{\Delta}} \sum_{k_1 k_2 k_3 k_4} : b_{k_1\uparrow}^\dagger b_{k_2\uparrow} b_{k_3\downarrow}^\dagger b_{k_4\downarrow} :, \end{aligned} \quad (10)$$

where $H_0(\psi) = \sum_{k\sigma} \epsilon_k \psi_{k\sigma}^\dagger \psi_{k\sigma}$ ($\psi = a^{(p)}, b$). This Hamiltonian describes the two last terms in the phase shift expansion Eq. (8).

The first term in Eq. (8), δ_0 , is incorporated into the definition of the b -particles in Eq. (10). These b -particles are single particle scattering states that describe an incoming s -wave suffering a scattering phase shift δ_0 at the boundary. Formally, to define the b -particles, one uses the unfolding transformation¹⁹ where $\psi_\sigma^{(s)}(x) = (1/2\pi) \int dk e^{-ikx} a_{k\sigma}^{(s)}$ ($x \in \{-\infty, \infty\}$) is a chiral field describing an s -wave scattering state with the left moving convention such that $x > 0$ is the incoming part and $x < 0$ the outgoing part, $x = 0$ being the boundary. From the definition of the phase shift δ_0 we have $\psi_\sigma^{(s)}(0^-) = e^{2i\delta_0} \psi_\sigma^{(s)}(0^+)$. We define the b -particles in terms of a scattering state with $\delta_0 = \pi/2$

$$\psi_\sigma^{(b)}(x) = \psi_\sigma^{(s)}(x) \text{sgn}(x), \quad (11)$$

and its Fourier modes $b_{k\sigma} = \int dx e^{ikx} \psi_{\sigma}^{(b)}(x)$.

Now consider $eV \neq 0$. As long as $eV \ll \tilde{\Delta}$ the system remains in the vicinity of the fixed point and the state at finite eV can be treated within the FL theory as a state with a non-thermal distribution of quasiparticles. We first consider single particle scattering states incoming from lead $i = L, R$. In second quantization those particles are annihilated by $(c_{k\sigma i})^{in}$. The occupation of those incoming waves is simply $\langle (c_{k\sigma i}^{\dagger})^{in} (c_{k'\sigma' i'}^{in}) \rangle = \delta_{kk'} \delta_{\sigma\sigma'} \delta_{ii'} f_i(\epsilon_k)$. Using Eq. (7), and the fact that before the scattering region ($x > 0$) the wave function of states $a_{k\sigma}^{(s)}$ and $b_{k\sigma}$ coincide, we have¹³

$$\begin{aligned} (c_{k\sigma L})^{in} &= (v_L b_{k\sigma} - v_R a_{k\sigma}^{(p)})/v, \\ (c_{k\sigma R})^{in} &= (v_R b_{k\sigma} + v_L a_{k\sigma}^{(p)})/v. \end{aligned}$$

This gives the nonequilibrium distribution function for the b -particles

$$\langle b_{k\sigma}^{\dagger} b_{k'\sigma'} \rangle = \delta_{kk'} \delta_{\sigma\sigma'} [\Gamma_L f_L(\epsilon_k) + \Gamma_R f_R(\epsilon_k)] / \Delta. \quad (12)$$

Since the occupation of the b -particles differs from the one defined at $T = eV = 0$, the second term of δH generates a constant elastic scattering $\frac{(R-1)n_{\bar{\sigma}}}{\pi\nu^2\tilde{\Delta}} \sum_{k_1 k_2 \sigma} b_{k_1\sigma}^{\dagger} b_{k_2\sigma}$, where

$$n_{\bar{\sigma}} = \sum_k \langle : b_{k\bar{\sigma}}^{\dagger} b_{k\bar{\sigma}} : \rangle = \nu(\Gamma_L \mu_L + \Gamma_R \mu_R) / \Delta. \quad (13)$$

As a result, the phase shift at energy ϵ relative to the

Fermi energy is given by [see Eq. (8)]

$$\delta_s(\epsilon) = \frac{\pi}{2} + \frac{\epsilon - \mu_K}{\tilde{\Delta}}, \quad (14)$$

where μ_K is a *nonequilibrium shift of the resonance*,

$$\mu_K = (R-1)(\Gamma_L \mu_L + \Gamma_R \mu_R) / \Delta. \quad (15)$$

This shift can be nonvanishing if $eV \neq 0$, and $U > 0$ (or equivalently $R \neq 1$). In the Kondo limit, $R = 2$, Eq. (15) implies that the resonance position μ_K shifts together with the average chemical potential $\bar{\mu} = (\mu_L + \mu_R)/2$: under $\bar{\mu} \rightarrow \bar{\mu} + \delta\mu$ at fixed eV , Eq. (15) gives $\mu_K \rightarrow \mu_K + \delta\mu$.

In order to calculate the current using the phenomenological Hamiltonian Eq. (10), one can use the Meir-Wingreen formula, and relate the Green's function $G^R(\epsilon)$ to the s -wave T -matrix, $T_s(\epsilon) = v G^R(\epsilon) v$. One can define a T -matrix, \tilde{T} , for the b -particles due to δH . It has an inelastic part denoted by \tilde{T}_{in} . The relation between T_s and \tilde{T} , accounting for the small inelastic term \tilde{T}_{in} , reads¹²

$$-\pi\nu T_s(\epsilon) = \frac{1}{2i} \left[e^{2i\delta_s(\epsilon)} - 1 \right] + e^{2i\delta_s(\epsilon)} \left[-\pi\nu \tilde{T}_{in}(\epsilon) \right]. \quad (16)$$

The leading contribution to \tilde{T}_{in} originates from the diagram shown in Fig. 1, containing three propagators of b -particles whose occupation is given by Eq. (12). We find, using the Keldysh technique,

$$\begin{aligned} -\pi\nu \text{Im} \tilde{T}_{in}(\epsilon) &= \frac{(R-1)^2}{4\tilde{\Delta}^2} \int d\epsilon_1 d\epsilon_2 d\epsilon_3 [1 + t(\epsilon_2)t(\epsilon_3) - t(\epsilon_1)\{t(\epsilon_2) + t(\epsilon_3)\}] \delta(\epsilon + \epsilon_1 - \epsilon_2 - \epsilon_3) \\ &= \frac{(R-1)^2}{2\tilde{\Delta}^2} [\pi^2 T^2 + (\epsilon - \kappa eV)^2 + (eV)^2 \frac{3A}{4}], \end{aligned} \quad (17)$$

where $t(\epsilon) = 1 - 2[\Gamma_L f_L(\epsilon) + \Gamma_R f_R(\epsilon)] / \Delta$ and $\kappa = (\Gamma_L \mu_L + \Gamma_R \mu_R) / [\Delta(\mu_L - \mu_R)] = (B\Gamma_L - \Gamma_R) / [(1+B)\Delta]$. Plugging Eqs. (14) and (17) into Eq. (16) and using the Meir-Wingreen formula Eq. (6), with $G^R(\epsilon) = v^{-2} T_s(\epsilon)$, we obtain the conductance in the scaling form of Eq. (1) with $G_0 = \frac{2e^2}{h} A$, $c_T = \frac{\pi^2 [1+2(R-1)^2]}{3}$, $T_K \rightarrow \tilde{\Delta}$ and

$$\alpha = \frac{9}{2\pi^2} \left(\kappa(R^2 - 1) \frac{1-B}{1+B} + \frac{2+(R-1)^2}{3} \frac{1+B^3}{(1+B)^3} + 3(R-1)^2 (\kappa^2 + \frac{1}{4}A) \right) / (1 + 2(R-1)^2). \quad (18)$$

The coefficient α can be expressed as a function of 3 independent variables such as the Wilson ratio R and the L-R asymmetry parameters A and B . In general, α depends on the the L-R asymmetry parameters however, in the strong coupling limit $U \rightarrow \infty$, equivalent to $R \rightarrow 2$, this dependence completely disappears from Eq. (18) and we obtain $\alpha[R=2] = \frac{3}{2\pi^2} = 0.1519$. In this limit, $\tilde{\Delta} \rightarrow 4\pi\sqrt{u/2\pi} \exp[-\pi^2 u/8 + 1/(2u)]$ which is the known expression for T_K , where $u = U/(\pi\Delta)$.

The value of α measured in QDs can be accounted for by charge fluctuations due to finite U since, for $R \neq 2$, Eq. (18) gives $\alpha[R, A, B]$ in the range $3/4\pi^2 = 0.0759 \leq \alpha \leq 0.3039 = 3/\pi^2$. Since one can tune the gate voltage in QDs and this corresponds to tuning ϵ_d , it is plausible that the SAM applies at one value of the gate voltage corresponding to maximal conductance. However the value $\alpha_{\text{SMT}} = 0.051 \pm 0.01$ measured in SMTs is lower than the expectation from the SAM. We note that a value

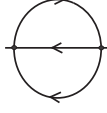


FIG. 1: Diagram used to calculate \tilde{T}_{in} . Each line is a b -particle propagator, with the nonequilibrium distribution Eq. (12).

$\alpha = 0.157 \pm 0.005$ was also measured²⁰ in Al/AlO_x/Sc planar tunnel junctions at low temperatures²¹ and can be accounted for in our theory.

We compare Eq. (18) to the results of other approaches for the KM and SAM. Firstly, our result is fully consistent with the results of Oguri⁸ based on Ward identities, however he concentrated on the special case of $A = B = 1$; Kaminski, Nazarov, and Glazman¹³ find $\alpha_{\text{KNG}} = \frac{3}{8\pi^2}$ for the KM for any A ; Konik, Saleur, and Ludwig⁷ find $\alpha_{\text{KSL}} = 4/\pi^2$ for the SAM for $A = 1$ and large U/Δ ; Pustilnik and Glazman¹² find for the KM for $A \ll 1$, $\alpha_{\text{PG}} = \frac{3}{2\pi^2}$; Rincón, Aligia, and Hallberg⁹ studied the SAM for the case $B = \Gamma_R/\Gamma_L$. A mistake was found in Eq. (10) in their paper, whereas the corrected formula²² $\frac{G}{G_0}|_{B=\frac{\Gamma_R}{\Gamma_L}} \simeq 1 - \frac{\pi^2(1+2(R-1)^2)}{3} \left(\frac{T}{\Delta}\right)^2 - \frac{4-3A+(2+3A)(R-1)^2}{4} \left(\frac{eV}{\Delta}\right)^2$ agrees with our Eq. (18). Given that α_{KSL} , which differs from our result, was obtained approximately and is not claimed to be exact, and given the agreement with the exact formulation due to Oguri,⁸ we are convinced of the

validity of our reported expression for α .

As another application of Eq. (10), one can calculate the shot noise $S = \int_{-\infty}^{\infty} dt \langle \delta I(t), \delta I \rangle$ for the SAM to leading order in $1/\tilde{\Delta}$ where $\delta I = I - \langle I \rangle$. At $T = 0$ and $A = B = 1$, using results for S based on the effective Hamiltonian δH of Eq. (10) with arbitrary coefficients,^{23,24} we obtain $S = \frac{4e^2}{h} \frac{1+9(R-1)^2}{12} \left(\frac{eV}{\Delta}\right)^2$. The ratio

$$e^* \equiv \frac{S}{2I_b} = \frac{1+9(R-1)^2}{1+5(R-1)^2} e, \quad (19)$$

where $I_b = \frac{2e^2}{h} V - I$, can be interpreted as a backscattering charge.²⁴ This charge crosses from $e^* = e$ for $R = 1$ (noninteracting resonance level) to $e^* = 5/3e$ for $R = 2$ (Kondo resonance).²³⁻²⁵

In conclusion, we extended Nozières Fermi liquid theory¹⁶ to account for charge fluctuations in the particle hole symmetric Anderson model and calculated the transport coefficient α present in the term describing the leading dependence on eV/T_K . We included explicitly the effects of L-R asymmetry of the device and discussed the relation to recent experimental results.^{14,15}

After this work was essentially completed, we became aware of another work²⁶ that obtains $\alpha = \frac{3}{2\pi^2}$ for the KM with arbitrary L-R asymmetry, consistent with our result in the special case without charge fluctuations.

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